Indian J. Phys. **76A** (1), 39-41 (2002)

IJP A
— an international journal

Crystal field and hyperfine effects in $ErNi_2B_2C$

Sulava Bhattacharyya
Department of Physics, Jadavpur University,
Kolkata-700 032, India

E-mail : sulava@juphys.ernet.in

Abstract : Recently, neutron diffraction and Mossbauer experiments have been performed on tetragonal $ErNi_2B_2C$. From these experiments, lowest crystal field energy gap, g-values and hyperfine splitting of ^{166}Er in the sample have been estimated. But no theoretical calculations have been done to explain these values. The present paper reports the results of detail crystal field (CF) calculation and effects of hyperfine interaction of ^{166}Er in the solid state $ErNi_2B_2C$. The CF state are eight Kramers' doublets of which $\left|\pm\frac{1}{2}\right\rangle$ is the ground one having highly anisotropic g-values. The observed hyperfine splitting was also explained by considering ^{166}Er in the ligand field environment.

Keywords : Crystal field, g-values, hyperfine splitting

PACS Nos. : 71.70.Ck, 78.20.Jx, 76.80.+y

1. Introduction

Recently, neutron diffraction [1] and Mossbauer experiment [2] have been done on tetragonal $ErNi_2B_2C$. The solid is paramagnetic at room temperature (RT) and antiferromagnetic at low temperature with $T_N = 6.8$K. At further low temperature, it becomes superconductor. Some physical properties of the sample have been estimated therein from the experimental data. The intensity of the most intense magnetic diffraction peak at $2\theta = 27.67°$ as a function of temperature, has been shown in the paper [1]. To fit the experimentally obtained curve, an empirical Brillouin function for $J = 15/2$, where J represents the total quantum number of the Er^{3+}, was used. The Mossbauer spectra [2] (MS) for ^{166}Er in $ErNi_2B_2C$ have shown that at RT no hyperfine (HF) splittings have been observed; but at about 5K, five finger spectra were observed.

From the experimental data, the crystal field (CF) energy difference between the lowest and the first excited levels was computed to be 9K. The g-values for the ground level were given as $g_{\parallel} = 1.2$ and $g_{\perp} = 9.6$. But no theoretical calculation was done therein to explain these values. Moreover, the paper [1] suggests that 'the crystal field effects are very likely important and the magnetic ground state is most probably a doublet with $J = \pm\frac{1}{2}$'.

To complement the experimental findings, the present paper reports the crystal field analysis on this tetragonal sample in paramagnetic region and explains the hyperfine effects of $^{166}ErNi_2B_2C$.

2. Theoretical considerations

A. *CF effects :*

Spectral [3] and magnetic studies [4] of several erbium compounds have indicated that the free ion ground term of Er^{3+} is $^4I_{15/2}$ followed successively by $^4I_{13/2}$, $^4I_{11/2}$ and $^4I_{9/2}$ at 6700cm^{-1}, 10400cm^{-1} and 12500cm^{-1} respectively above the ground one. The free ion levels of Er^{3+} are not pure Russel-Saunders (RS) states, but are admixtures of different RS states. The lowest intermediate coupled free ion state in Er^{3+} is given below [5] :

$$\left|I_{15/2}\right\rangle = 0.9838\left|{}^4_3 I_{20}\right\rangle - 0.1783\left|{}^2_3 K_{21}\right\rangle - 0.0191\left|{}^2_3 L_{21}\right\rangle. \quad (1)$$

For the present system, the sample is body-centred tetragonal in structure (Figure 1) with an additional carbon atom in each erbium layer. The CF potential on Er^{3+} in this sample is

$$V_{CF} = B_2^0 V_2^0 + B_4^0 V_4^0 + B_6^0 V_6^0 + B_4^4\left(V_4^4 + V_4^{-4}\right) + B_6^4\left(V_6^4 + V_6^{-4}\right), \quad (2)$$

where B_k^q and V_k^q are respectively the even parity CF parameters (CPF) and the one electron intra-configuration unit tensor operators [6].

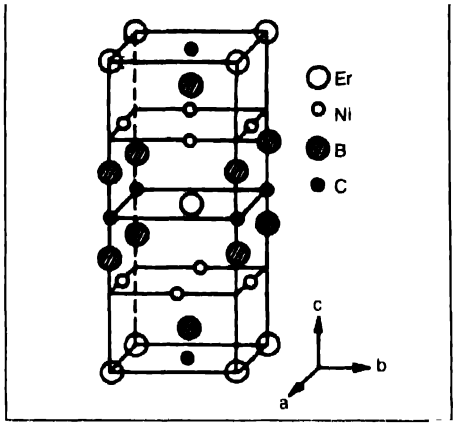

Figure 1. Structure of body centred tetragonal $ErNi_2B_2C$

Operating with V_{CF} (eq. (2)) on intermediate coupled free ion state of Er^{3+} (eq. (1)), the CF energy levels have been determined. By fitting the energy gap between the ground and the first excited CF levels and the magnetic splitting factors of the ground state, the accurate set of CPF were determined. The g-values of the ground CF state were obtained from the relations :

$$g_i = 2g_J \left\langle \Psi_g | J_i | \Psi_g \right\rangle, \tag{3}$$

where J_i is J_z for $g_{\parallel}$ and J_x for $g_{\perp}$ respectively, and g_J is the Lande' splitting factor.

B. HF effects :

The hyperfine Hamiltonian for this rare-earth compound is :

$$H_{hf} = \left[AS_z I_z + B(S_x I_x + S_y I_y)\right] + \frac{e^2 Q}{4I(2I-1)} \langle q_{zz} \rangle_T \left[3I_z^2 - I(I+1)\right]. \tag{4}$$

The terms inside the first square bracket are due to nuclear magnetic HF interaction, A and B being HF constants and S represents the effective electronic spin. The crystalline electric field interaction between the central Er^{3+} cation and the ligands produces an electric field gradient (EFG) at the nucleus. The second term of eq. (4) is the product of the EFG and the quadrupole moment Q of the nucleus. The EFG has two main parts arising from lattice and 4*f*-electronic contribution [7].

^{166}Er has zero nuclear spin ($I_g = 0$) at ground state and consequently, it has no hyperfine splitting. For first excited state, which is situated at about 80.6 kev above the ground one, the spin value is two ($I_e = 2$). Hence, this level ought to show HF splittings. Operating by H_{hf} on the basis state $|m_s, m_I\rangle$, ten hyperfine levels are obtained. Mossbauer lines were obtained by considering the dipolar transitions, between the HF split levels of I_e and I_g, for which the selection rule is $\Delta m_I = 0, \pm 1$ The width of the HF structure was estimated from these transition lines.

3. Results and discussion

For Er^{3+}, which is a $J = 15/2$ system, eight Kramers' doublets were obtained under CF perturbation. The best fitted CF energy levels are shown in Figure 2, and the lowest level has $\mu = 1/2$

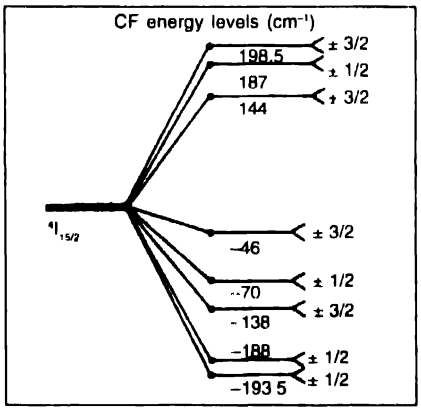

Figure 2 Crystal field energy levels of $ErNi_2B_2C$ (not to scale)

Table 1. Some CF properties of $ErNi_2B_2C$

CF parameters (cm^{-1})	Lowest two CF levels (cm^{-1})	$\Delta = E_2 - E_1$ (K)	Lowest two CF wavefunctions	g-values (calculated and observed)
$B_2^0 = -50 \pm 15$		8 [present work]	$\Psi_g = 0.19268 \mid \pm 15/2 > - 0.57303 \mid \pm 7/2 > - 0.50294 \times \mid \mp 1/2 \rangle - 0.61771 \mid \mp 9/2 \rangle$	$g_\parallel = 1.3$ $g_\perp = 10.82$
$B_4^0 = 60 \pm 3$				[present work]
$B_6^0 = -64 \pm 2$	$E_1 = -193.5$			
$B_4^4 = -270 \pm 6$	$E_2 = -188$	9 [Ref 1]	$\mid \psi_g' \rangle = 0.18635 \mid \pm 15/2 \rangle - 0.55623 \mid \pm 7/2 > - 0.51406 \times \mid \mp 1/2 \rangle - 0.6258 \mid \mp 9/2 \rangle$	$g_\parallel = 1.2$ $g_\perp = 9.6$
$B_6^4 = 210 \pm 5$				[Ref 2]

where μ is the crystal quantum number. The CFP and CF wavefunctions for the lowest two levels are shown in Table 1. This table indicates that this set of CFP explains the observed lowest energy gap and g-values for $ErNi_2B_2C$ very well.

It was found [4,7] that the values of $A/g_{\parallel}$ and $B/g_{\perp}$ for some erbium compounds remain constant. Taking the average values of these quantities and substituting the corresponding g-values (for the ground state), the hyperfine constants for this sample have been calculated. These values are $A = 44.7 \times 10^{-4}$ cm^{-1} and $B = 379.7 \times 10^{-4}$ cm^{-1}.

It has been mentioned earlier that the ground nuclear state has no HF splitting since I_g has zero value. The first excited state ($I_e = 2$) should have HF splittings. To get the required HF energy levels, the electric quadrupole interaction parameter ($P_{ee} = \frac{e^2Q}{4I(2I-1)} \times \langle q_{zz} \rangle_T$) for the first excited state was calculated. The values of necessary physical constants were taken from earlier work [8]. The value of P_{ee} was found to be 3.055×10^{-4} cm^{-1}.

The thermal variation of variable part of EFG due to 4*f*-electronic contribution is shown in Figure 3. The figure indicates that at about 3K, its value becomes constant.

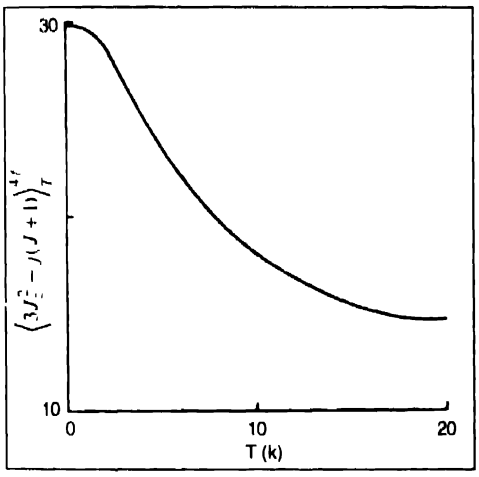

Figure 3. Thermal variation of $\langle 3J_z^2 - J(J+1) \rangle_T^{4f}$ (4*f*-electronic contribution of EFG)

Operating with the corresponding H_{hf} on $|m_S, m_I>$ state ten HF levels were obtained for $I_e = 2$. Out of these ten levels one is doublet and eight are singlets (Table 2).

Table 2. HF levels of ^{166}Er in $ErNi_2B_2C$ (*S* represents singlet and *D* represents doublet).

Nuclear term	A, B, P_{ee} (10^{-4} cm^{-1})	HF levels (10^{-4} cm^{-1})
$I_e = 2$	A = 44.7	- 2408.5 (S)
		-1896.4 (S)
		- 1307.7 (S)
	B = 379.7	- 743.8 (S)
		- 26.4 (D)
		775.5 (S)
	P_{ee} = 3.055	1323.2 (S)
		1823.6 (S)
		2395.3 (S)

Total width of MS at and below 3K was found to be 17cm/sec which is very close to that of observed spectra [2].

Hence we can conclude that the given set of CPF for $ErNi_2B_2C$ explains the CF properties, g-values as well as the HF splitting of the system.

References

[1] S K Sinha, J W Lynn, T E Grigereit, Z Hossain, L C Gupta, R Nagarajan and C Godart *Phys. Rev.* **B51** 681 (1995)

[2] E Alleno, P Benville, C Godart, L C Gupta, G Hilscher, J A Hedges, Z Hossain, H Micher, R Nagarajan and C Vaast, *Hyp Int* **C1** 171 (1996)

[3] H Dieke *Spectra of Energy Levels of Rare-Earth Ions in Crystals* (New York Wiley) (1968)

[4] S Dasgupta, M Saha, S Mroczkowski and D Ghosh *Phys Rev.* **B27** 6960 (1983)

[5] S Hufner *Optical Spectra of Transparent Rare-Earth Compounds* (New York Academic) (1978)

[6] G Racah *Phys Rev* **62** 438 (1942)

[7] S Dasgupta and D Ghosh *J Appl Phys* **63** 5835 (1988)

[8] J Pelzl, S Hufner and S Scheller Z *Phys* **231** 377 (1970)